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Crossover from classical to quantum regime in Ce-lattice systems ☆

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Abstract

Different types of magnetic phase diagrams are analyzed with the scope to detect signs of a crossover between the classical (thermal fluctuations) regime and the critical (quantum fluctuations) one. In systems where $T_{N,C}$ is driven down to the mK range by alloying Ce-ligands (x), clear changes in the magnetic phase are observed once $T_{N,C}<2\,K$. On the contrary, when T_N vanishes at finite temperature (driven by pressure or x) the phase boundaries do not reach temperatures lower than $\approx 3\,K$. In a third type of phase diagrams, characterized by a weak $T_N(x,p)$ dependence, signs of phase separation are suspected. Distinctive properties of each mentioned group are recognized and some exemplary cases analyzed in detail. Those properties coincide indicating a crossover temperature slightly above 2 K.

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A quantum critical point is defined as the end point of a phase transition driven to zero by a non-thermal control parameter [1] where it becomes a quantum phase transition. The usual control parameters are chemical potential (realized by e.g. alloying the Ce-ligands 'x') or pressure 'p', which reduces the ordering temperature $T_{N,C}$ by weakening the RKKY magnetic interaction whereas the Kondo screening is enhanced [2]. While at finite temperature the transitions are dominated by thermal fluctuations, by approaching $T=0$ the quantum fluctuations become dominant since their energy is not thermal dependent. Hence, a crossover between those regimes is expected as $T_{N,C}(x,p)$ decreases. The aim of this work is to search for signs of such a crossover between the temperature ranges where each type of fluctuations are dominant. For that goal, distinctive characteristics of various phase diagrams of Ce-lattice compounds are investigated.

As it was established some years ago [3], three types of phase diagrams can be distinguished. One includes the systems whose $T_{N,C}(x)\to 0$ monotonously as $x\to x_{cr}$ after being traced down to at least 10% of its original value at $x=0$ (see Fig. 1a). Another corresponds to those compounds where the phase boundary vanishes at finite temperature, i.e. T_N cannot be traced beyond certain value of 'x' or 'p' (see Fig. 1b). The third type contains the cases displayed in Fig. 2, where T_N is practically independent of the control parameters before vanishing, also at finite T. More recent phase diagrams (see e.g. Ref. [4]) confirm that classification and allow to search for distinctive characteristics of each type of phase diagrams.

After such analysis one recognizes that those belonging to the first group are characterized by: (i) showing the maximum $T_{N,C}(x)$ range, with the lowest reported ordering temperature for $CePd_{0.13}Rh_{0.87}$ at $T_C(x)=27\,mK$ [5]. (ii) All these phase diagrams show a change of curvature at a characteristic concentration x^*, which corresponds to similar temperature values: $T^*(x^*)\approx 2\,K$ [6]. (iii) The maximum value of their specific heat divided by T: C_{max}/T, is nearly constant within the $x^*\leqslant x\leqslant x_{cr}$ range [7]. These properties allow to identify a *pre-critical* region [6] on the magnetic side of the critical point, not detected in the other two types of phase diagrams.

It is evident that the onset of the pre-critical region plays a relevant role in the evolution of the ground state

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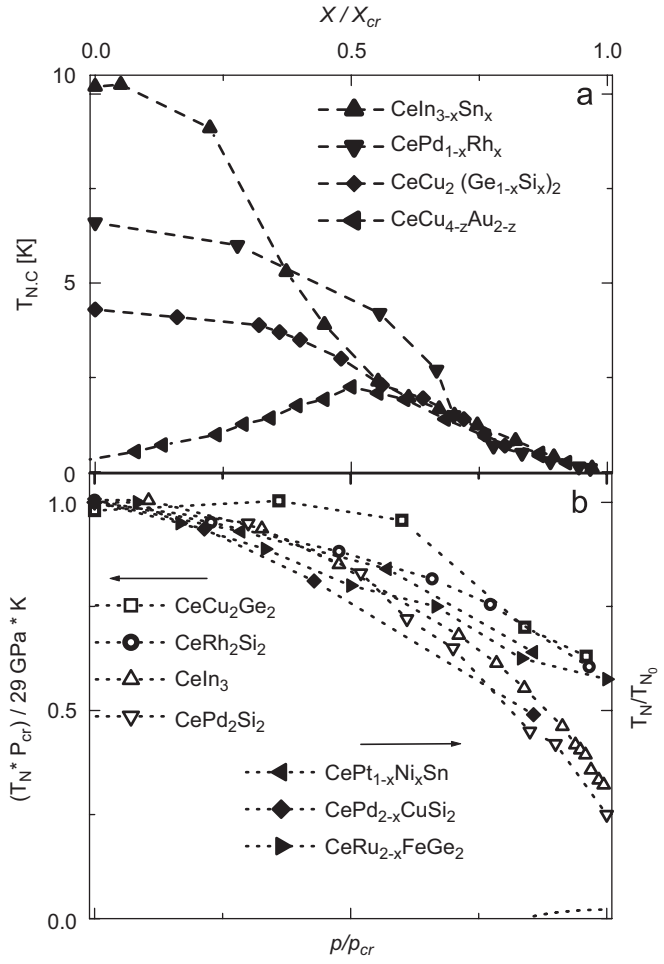

Fig. 1. Two types of magnetic phase boundaries. (a) Systems belonging to first group (type-I) and (b) to the second (type-II). Within the latter group, those driven by pressure (alloying) are referred to the left (right) axis. The normalization procedure is described in the text. For the references see Ref. [4].

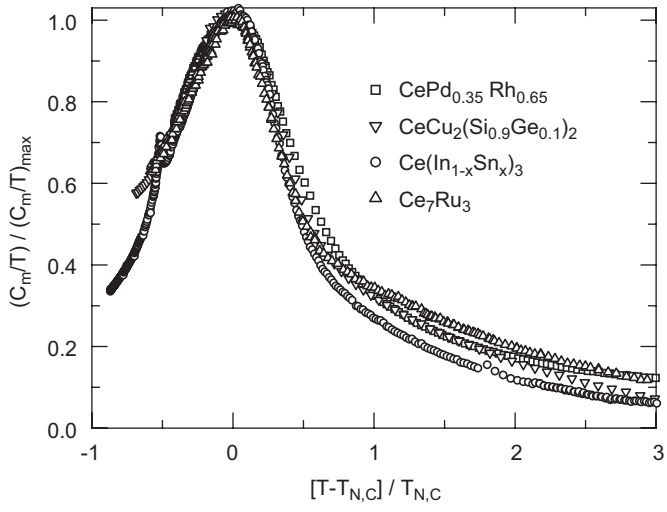

Fig. 2. Comparison of normalized specific heat jumps at their respective transitions for type-I systems at $x = x^*$. Stoichiometric Ce_7Ru_3 [11] is included for comparison.

magnetic properties of the alloyed systems. The fact that in early phase diagram determinations, performed at $T > T^*$, the $T_{N,C} \to 0$ extrapolations coincide with the present x^* values suggests that two ranges of thermal energy: $T > T^*$ and $T < T^*$, can be distinguished. Then, T^* represents a sort of energy crossover between two regimes, the classical one controlled by canonical thermal fluctuations and that of lower energy dominated by quantum fluctuations. Since the respective order-parameters (OP) are not yet established in those regions, the question whether they belong to the same magnetic phase remains open. A preliminary evidence in that sense is given by the frequency dependence study of the AC-susceptibility of $CePd_{1-x}Rh_x$ which shows no dependence between $0 < x < x^*$, while it occurs for $x > x^*$ [5,7].

Many properties of alloyed systems are suspected to be due to *disorder effects* [4]. However, no distinction between different origins, nature and effectiveness of disorder are usually taken into account. Obviously, to dope the Ce-lattice destroys the coherence of the lattice, whereas by alloying Ce-ligands only a local distortion is produced. Even in the latter case a distinction between structural and electronic effects (e.g. atomic size difference of Ce-ligands and sign of their electronic potential) has to be done. One cannot expect the same strength of disorder effect between atoms of similar size (e.g. Pd/Rh or In/Sn) than between those of different size (e.g. Pd/Ni or Cu/Au). Furthermore, a stronger disorder effect is expected by alloying *electron-like* atoms with *hole-like* ones (e.g. Cu/TM in $UCu_{5-x}TM_x$, TM = transition metals [4] or Pd/Ag in Ce(Pd,Ag) [8]) due to the different signs of their Coulomb potential than between those of the same sign (e.g. Pd/Rh or In/Sn). On the other hand, atomic disorder is expected to rise progressively with x and to extend beyond any specific concentration till decreasing once $x \to 1$. However, if x^* marks a crossover between two different regimes, signs of an *intrinsic* disorder can be expected at that concentration. Effectively, some features currently associated to disorder only occur at the vicinity of x^*. The systems included in Fig. 1a behave as in this case, showing a specific heat transition that broadens just close to x^* and then changes into a Z shape within the *pre-critical* region [6]. This behavior is observed independently of the absolute value of x^*, which occurs between $x = 0.10$ in $Ce(In_{1-x}Sn_x)_3$ [9] and $x = 0.60$ in $CePd_{1-x}Rh_x$ [10].

Despite such a large difference in their relative ligand concentrations at $x = x^*$, the respective specific heat transitions are remarkably similar as it is shown in Fig. 2. In order to compare different systems, we have scaled the temperature as $[T - T_{N,C}]/T_{N,C}$ and normalized the specific heat C_m/T to its maximum value $(C_m/T)_{max}$. The so-called Λ shape of those transitions indicate that there is similar entropy contribution above and below $T_{N,C}$ like predicted for the 2d-Heisenberg model [7]. Since in this group $(C_m/T)_{max}$ is nearly constant, from $\Delta C_m(T_N) \propto T_N^{(d-1)}$ [12] this observation coincides with a low dimensionality behavior. Though disorder due to random alloying effects cannot be discarded,

it is unlikely to find such an ample coincidence among different systems and concentrations.

Examples of Ce-lattice phase diagrams belonging to the second group (type-II) are presented in Fig. 1b, including x and p driven cases. These phase boundaries vanish at finite temperature, notably at $T > 3$ K. All p driven compounds included in Fig. 1b show a superconductive phase at their respective critical pressure p_{cr}. Their respective phase boundaries can be normalized as $(T_N \times p_{cr})/29$ GPa K, as shown on the left axis of Fig. 1b. This normalization leads to the empirical observation that $T_N * p_{cr} \approx 29 \pm 2$ GPa K for all Ce compounds becoming superconductors under pressure. Notice that, like in type-I, no superconductivity is observed in the alloyed systems of this group.

The only exception to the $T \approx 3$ K threshold is observed in the p driven $CePd_2Si_2$ [13]. In this compound, $T_N(p)$ is traced back as a kink in resistivity $\rho(T,p)$ measurements. The quality of those measurements allow to evaluate the temperature derivative $\delta\rho/\delta T$, which shows a replica of the specific heat jump at T_N as displayed in Fig. 3 after Ref. [13]. Assuming $\delta\rho/\delta T \propto C_m$, it is possible to evaluate the evolution of the degrees of freedom involved in the magnetic transition as a function of pressure by computing $\Delta(\delta\rho/\delta T) \propto \Delta C_m(T_N)$. To test whether the phase boundary of this compound really vanishes at finite temperature or the lack of its detection below 2 K is due to experimental limitations, one can search at which temperature (either finite or zero) the $\Delta(\delta\rho/\delta T) \to 0$ extrapolation corresponds to. As shown in the inset of Fig. 3, such an extrapolation corresponds to $T \approx 2$ K. This proves that the degrees of freedom related to the transition vanish at finite temperature and no transition can be expected below that temperature.

Some phase diagrams of systems showing a weak $T_N(x,p)$ dependence before vanishing at finite temperature (type-III) are displayed in Fig. 4. There is a clear difference in these examples between those with x or p dependence.

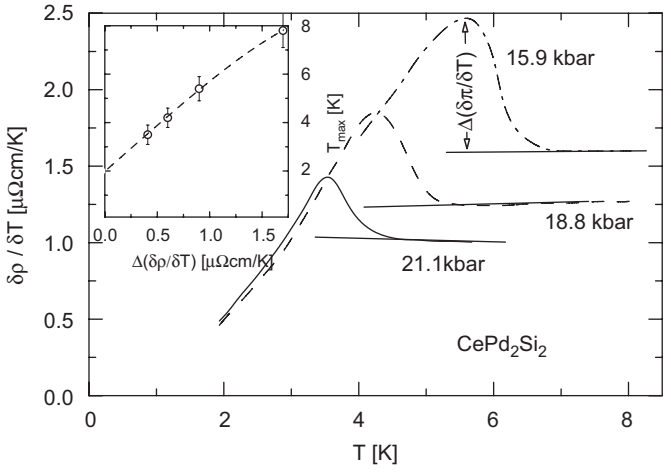

Fig. 3. Thermal variation of the resistivity derivative under high pressure (after Ref. [13]). Inset: extrapolation of the temperature of the maximum of $\delta\rho/\delta T$ to the zero value of $\Delta(\delta\rho/\delta T)$.

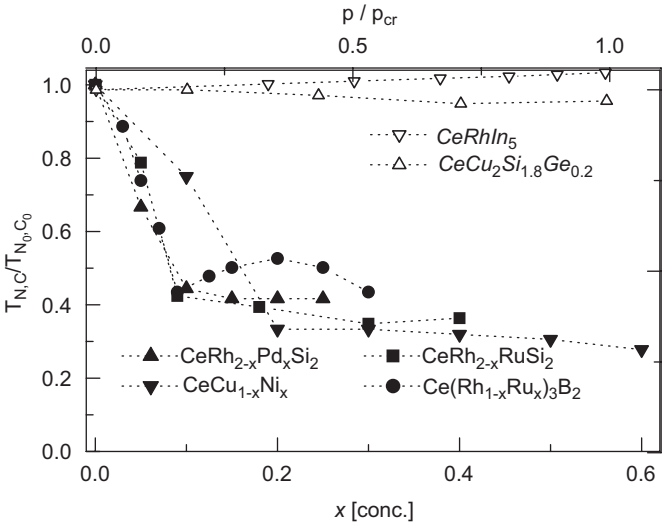

Fig. 4. Type-III magnetic phase boundaries, showing p (open) and x (full symbols) independent behavior. The latter showing a plateau before vanishing. Respective references are given in Ref. [4].

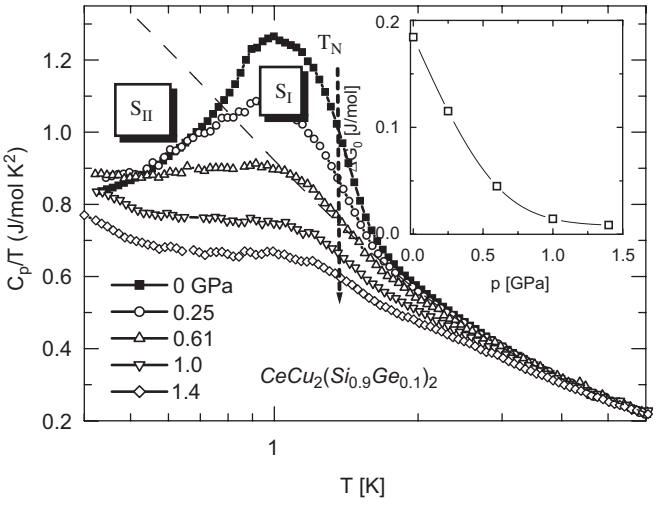

Fig. 5. Specific heat data of a type-III compound taken from Ref. [14]. Dashed curve indicates the reference function C_{Ref}/T respect of which $S_I = S_{II}$. Inset: analysis of the free energy gain (ΔG) as a function of pressure.

While in the former $T_{N,C}(x)$ drops down before entering into a plateau, in the latter $T_N(p)$ practically does not change with respect to its value at $p = 0$. Notably, the plateau observed in the alloyed systems has a similar *origin/plateau* ≈ 0.35 ratio despite their very different $T_{N,C}(x = 0)$ values. In these systems the question arises whether the phase boundary ends in a first-order transition.

Such a possibility is analyzed taking profit of the specific heat results from $CeCu_2(Si_{0.9}Si_{0.1})_2$ [14] shown in Fig. 5, following the procedure proposed in Ref. [3]. The entropy condensed by ordering, ΔS_{MO}, is evaluated as the difference between the measured entropy gain $S_m = \int C_p/T\,dT$ and that of a reference function $S_{Ref} = \int C_{Ref}/T$, being C_{Ref}/T the function with respect to which the entropy is compensated below T_N, i.e. $S_I = S_{II}$ as shown in Fig. 5. This leads

to $\Delta S_{MO} = S_{Ref} - S_m$. The function C_{Ref}/T is obtained as an extrapolation of C_p/T from $T > T_N$, using a heuristic function $\gamma_T = \gamma_0/(T^r + E)^s$ [6]. The function fulfilling the $S_I = S_{II}$ condition in this case is: $C_{Ref}/T = \gamma_T = 1.13/(T^{1.6} + E(p))^{0.55}$. We notice that E is the only p dependent parameter, whereas γ_0, r and s are independent of p (within $\approx 5\%$ dispersion). The pressure variation of $E(p)$ is described by $E = 0.4 + 0.94(\pm 0.1)p^2$. The free energy gained by ordering, $\Delta G(T)$, is then evaluated integrating the entropy difference as $\Delta G = \int \Delta S_{MO}\, dT$. The ΔG value at $T = 0$, ΔG_0, represents the total energy gained. The inset in Fig. 5 shows that $\Delta G_0(p)$ tends to zero asymptotically as $T_N(p)$ vanishes, without any sign of discontinuity. This observation contradicts the possibility of a first-order transition at p_{cr} because in this case $T_N(p)$ vanishes by exhausting the magnetic degrees of freedom, whereas the magnetic interaction ($J_{RKKY} \propto T_N$) remains practically unchanged. This behavior resembles that of a phase separation scenario since the *intensive* parameter J_{RKKY} remains constant while the *extensive* one (i.e. number of interacting moments involved) decreases.

It is predicted that, in general, disorder decreases the critical temperature from its *clean* value (cf. T_C^0) [1]. Within that range of temperature, the system does not show global order but 'rare regions' of static local order of arbitrary size. Since they are finite and coupled among them, no true static order develops at large scales but a slow fluctuating OP [1]. Then, with decreasing temperature the apparent critical point shifts towards the disordered phase, with rounded transitions [1]. Though this model is restricted to Ising OP symmetry, we can test up to which extent it is applicable to the systems here investigated.

By comparing the distinctive properties of each type of phase diagram, one sees that these model predictions are mainly realized in the type-I group. There, alloying effects may introduce some degree of local disorder as it was discussed before. Strictly, $CePd_{1-x}Rh_x$ is the unique ferromagnet reported, nevertheless the distinctive properties of this group extend to the anti-ferromagnetic members. If $T^*(x^*)$ is identified with T_C^0, one observes that the critical point shifts towards the disordered phase as the temperature decreases according to the model. The effects of the fluctuating OP are detected by χ_{AC} measurements performed on $CePd_{1-x}Rh_x$ [5,7]. On the other hand, the rounded transitions are observed around $x = x^*$ as depicted in Fig. 2.

The end point at finite temperature of the systems included in type-II phase diagrams suggests that the long-range OP collapses without forming 'rare regions'. This is more likely expected in stoichiometric compounds driven by pressure with an homogeneous OP due to the absence of local disorder. Coincidentally in those compounds a superconductive phase appears at lower temperature [4].

In the case of type-III phase diagrams, we have seen that the exemplary system $CeCu_2(Si_{0.9}Si_{0.1})_2$ shows exhausting magnetic degrees of freedom where $T_N(p)$ vanishes. This property may coincide with the formation of the mentioned 'rare regions'. Nevertheless, since $T_{N,C}(x,p)$ is nearly constant an intrinsic difference arises with respect to type-I systems regarding the RKKY interaction dependence on x and p. Then, while in type-I cases the critical point is approached by lowering $T_{N,C}(x)$, the behavior of type-III phase boundaries is more likely associated to a phase separation scenario.

We conclude that the distinctive characteristics observed in different types of Ce-lattice phase diagrams correspond to intrinsic differences in the properties of the respective systems. By comparing their behaviors a crossover between high (thermal) and low (quantum) energy regimes can be proposed at around $T^* \approx 2\,K$, where the onset of a *pre-critical* region is observed in the type-I group. Whether this is a different phase with respect to that of $0 < x < x^*$ remains an open question, though the model predictions only apply to the $x > x^*$ range.

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